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THE ATMOSPHERIC AEROSOL

L. DUFOUR

Translated by Glenn B. Hoidale
March 1969

ATMOSPHERIC SCIENCES LABORATORY
WHITE SANDS MISSILE RANGE, NEW MEXICO

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1. Introduction

The atmospheric aerosol may be defined as humid air in which small liquid or solid particles are more or less irregularly dispersed, the largest of these particles being sufficiently small to remain in suspension in the atmosphere for a certain time. The liquid or solid particles consisting of clouds, precipitation and of certain atmospheric phenomena are only considered to constitute part of this aerosol.

The solid or liquid particles contained in the atmospheric aerosol are generally referred to as "condensation nuclei". This appellation is fundamentally correct since all particles of the atmospheric aerosol, whatever their physical or chemical properties might be, can play the role of condensation nuclei, if the air is sufficiently supersaturated with water vapor. However, since the relative humidity* of the atmosphere rarely exceeds 101%, only a part of these particles are truly condensation nuclei.

*Remember that relative humidity, indicator of the humidity of the air often used in meteorology, is defined by the formula

$$u = 100(P_v / P_{v_{\infty}}^*)$$

where P_v is the partial pressure of water vapor and $P_{v_{\infty}}^*$ the saturation vapor pressure, in the presence of a plane surface of pure water at the same pressure and the same temperature as the air.

When the air is saturated with respect to a plane surface of pure water ($P_v = P_{v_{\infty}}^*$), the relative humidity is equal to 100%. When the air is supersaturated with respect to a plane surface of pure water ($P_v > P_{v_{\infty}}^*$), the humidity is greater than 100%; when the air is not saturated with respect to such a surface ($P_v < P_{v_{\infty}}^*$), the relative humidity is less than 100%.

This is why we only use the expression "condensation nuclei" in the very precise case of those nuclei about which the droplets form. In order to distinguish the particles of the atmospheric aerosol, we simply use the term "nuclei" because it has been established by usage. However, it sometimes can lead to confusion.

The radii* of atmospheric nuclei extend from about $5 \times 10^{-3} \mu$ to 20μ ($1 \mu = 10^{-4} \text{ cm}$); therefore, the spectrum of radii of atmospheric nuclei, expressed in μ , extends over nearly four orders of magnitude. These limits are somewhat arbitrary, especially the upper limit. It has been selected because nuclei having a radius greater than 20μ can only remain in suspension in the atmosphere for a limited time, due to the effect of sedimentation (see 4.1.3.); thus they can only be observed near the source which gave birth to them. As for the lower limit of $5 \times 10^{-3} \mu$, it arises because nuclei having radii smaller than this dimension have an ephemeral existence; as a matter of fact they are rapidly captured by the larger nuclei through the effect of Brownian motion (see 4.2.4.). By way of comparison, let us recall that the radius of a molecule is, in general, between $10^{-3} \mu$ and $10^{-4} \mu$.

*The notion of dimension or a nucleus is still more ambiguous than that of radius of a nucleus. Numerous definitions, which we won't take up, have been proposed to rectify this ambiguity. In our case, we shall consider an average radius.

In the case of the atmospheric aerosol one studies, in general, classes of nuclei. Actually, according to Junge (1952), one distinguishes three classes of nuclei: Aitken nuclei ($r \leq 0.1 \mu$), large nuclei ($0.1 \mu < r \leq 1 \mu$) and giant nuclei ($r > 1 \mu$).

2. Granulometry of nuclei

2.1. Granulometry near the ground

2.1.1. General

According to Junge (1963b), the average distribution over the continent and over the ocean of the volume concentration** n of the nuclei as a function of their radius r takes the form indicated in Fig. 1. This figure is sufficiently clear that interpretation is

CAPTION Figure 1. Average distribution over the continent (Curve C) and over the ocean (Curve O) of the concentration n of atmospheric nuclei as a function of their radius r (Junge, 1963b)

**When the type of concentration is not specified, it is understood that it refers to a volume concentration.

not necessary. One salient feature of the distribution over the continent is the straight line portion between 0.1μ and 10μ from which it follows that in this interval the relation between the radius r and the concentration n of the nuclei follows a law of the form

$$n = Cr^{-3} \quad (1)$$

where C is a constant and where $\lambda=3$. If the nuclei are assumed to be spherical, it follows that in the interval under consideration, the total volume, V , of the nuclei of a given radius is constant for a unit volume of air since

$$V = n \frac{4}{3} \pi r^3 = \frac{4}{3} \pi C \quad (2)$$

2.1.2. Aitken nuclei

These nuclei are called Aitken because their concentration can be easily determined with the aid of an Aitken nuclei counter. The principle of this instrument is as follows: a given volume of saturated atmospheric air is suddenly expanded; the resultant supersaturation is determined so that droplets are formed around those nuclei having diameters greater than about $5 \times 10^{-3}\mu$; then the number of droplets formed are counted and divided by the volume of the sample of air to obtain the Aitken nuclei concentration. This number, in reality, gives a measure of the total concentration of atmospheric nuclei with the exception of that portion due to small ions. However, noting the relatively low concentration of large nuclei and giant nuclei compared to that of Aitken nuclei (see Fig. 1), it may be assumed that the measured values correspond to Aitken nuclei.

The small ions, to which we have already referred, are produced by cosmic rays, radioactive radiation, electrical discharges, etc., which serve to detach an electron from a molecule causing the formation of a positive ion. The liberated electrons rapidly attach themselves to other molecules to form negative ions.

The ions can become attached to atmospheric nuclei to form large ions, which in turn may be subdivided into several groups according to radius (see, for example, Israel and Schulz, 1932):

Large medium ions	$8 \times 10^{-3} \mu \leq r < 2.6 \times 10^{-2} \mu$
Langevin ions	$2.6 \times 10^{-2} \mu \leq r < 5.5 \times 10^{-2} \mu$
Ultralarge ions	$r \geq 5.5 \times 10^{-2} \mu$

The atmospheric ions, once formed, behave like nuclei and participate, for example, in Brownian motion. Yet the small ions aren't considered part of the atmospheric aerosol because they cease to exist when their charge is neutralized, which is different from large ions which continue to exist as Aitken nuclei when this phenomenon occurs. Noting the small size ($6 \times 10^{-4} \mu$ mean) and the relatively low concentration (several hundred per cm^3) of the small ions, they can be neglected in cloud physics but not in atmospheric electricity where their role is fundamental.

The concentration of Aitken nuclei near the ground varies markedly in time and space (see, for example, Landsberg, 1938; Burckhart and Flohn, 1939). For a long time these variations gave the impression that Aitken nuclei play a fundamental role in the formation of clouds and precipitation. This belief started research on this subject in the wrong direction, and it is for this reason that the roles played by large nuclei in the formation of cloud droplets and by the giant nuclei in the formation of precipitation have been realized so late.

Table 1, due to Landsberg (1938), gives an idea of the variation in concentration of Aitken nuclei as a function of the place of observation. For example, from this table it is seen that the mean concentration is much greater in large cities than over the ocean. Also, it is seen that the minimum concentration can be very low in unpolluted regions which has been confirmed by Fenn (1960) who observed concentrations of several nuclei per cm^3 in Greenland.

TABLE 1. Concentration per cm^3 of Aitken nuclei at various locations

Location	Number of sites	Number of observations	Mean	Maximum Mean	Minimum	Maximum Absolute	Minimum
Large city	28	2,500	147,000	379,000	49,000	4,000,000	3,500
City	15	4,700	34,300	114,000	5,900	400,000	620
Open Country	25	3,500	9,500	66,500	1,050	336,000	180
Sea coast	21	2,700	9,500	33,400	1,560	150,000	0
Island	7	480	9,200	43,600	460	109,000	80
Ocean	21	600	940	4,860	840	39,800	2

The result of this variation as a function of the place of observation is that Aitken nuclei are unquestionably of continental or artificial origin.

The size distribution of Aitken nuclei can be determined by measuring their diffusion coefficient (see, for example, Nolan and Doherty, 1950) or

by measuring their mobility in an electrical field (see, for example, Israël and Schulz, 1932). It can also be determined, as done by Linker (1943) and Hosler (1950), by capturing the droplets formed in an Aitken counter on a reticule and observing the residue under an electron microscope.

Measurements based on the mobility of Aitken nuclei in an electrical field (see, for example, Israël and Schulz, 1932; Junge, 1955) indicate that these nuclei are grouped about certain preferred radii. They also show that the lower limit of the radius of these nuclei varies from one day to the next and is on the average $4 \times 10^{-3} \mu$. As for the radius corresponding to the maximum concentration of these nuclei, it varies between 10^{-2} and $10^{-1} \mu$, its most frequent value being in the neighborhood of $3 \times 10^{-2} \mu$.

Recent measurements on the granulometry of Aitken nuclei have been carried out by Twomey and Severynse (1964) with the aid of a method whereby measurement is made of the total concentration of nuclei at various stages of their diffusional decay. These authors have shown that the variation of the concentration of nuclei of radius from $5 \times 10^{-3} \mu$ to $2 \times 10^{-1} \mu$ is not continuous. There is a sharp minimum, corresponding to an absence of nuclei, in the vicinity of $r = 1.5 \times 10^{-2} \mu$ and a relatively stable and well marked maximum through $r = 6 \times 10^{-2} \mu$, a value slightly greater than that of $3 \times 10^{-2} \mu$ found by Junge (1955).

No farther emphasis will be placed on the secondary role of Aitken nuclei: in atmospheric optics; in atmospheric chemistry because their mass represents a maximum of 10 to 20% of the total mass of the nuclei (Junge, 1963b) and also in microphysics of clouds because the atmospheric droplets (clouds, fog) are normally formed on large and giant nuclei, the exception being around large Aitken nuclei.

2.1.3. Large nuclei

There are several methods for studying the granulometry of these nuclei: capture by spider web (see, for example, Dessens, 1946, 1949), capture by impaction on an obstacle (conimeter) (see, for example, May, 1945; Junge, 1953), thermal precipitation (see, for example, Watson, 1936), etc. Obtaining samples by these methods offers no particular difficulties, but the analysis of these samples is long and tedious. The analysis is particularly difficult for the small radii, the limit of resolution for the optical microscope being about 0.3μ and measurements with the electron microscope being inexact because of the evaporation of volatile substances from the nuclei resulting from the increase in temperature due to electron bombardment. A recently developed instrument, the Goetz aerosol spectrophotometer (1957) (see also Goetz and Preining, 1960; Goetz and Kallai, 1964) based on the principle of

centrifugal separation, allows one to obtain reliable data on the concentration of nuclei in the radius range of 0.01μ to 1μ .

Over the continent, the average concentration of large nuclei varies generally between 10^{-4} per cm^3 for $r = 0.1\mu$ and 10 per cm^3 for $r = 1\mu$ (see Fig. 1, Curve C). Over the ocean the variation is less marked and more irregular (see Fig. 1, Curve D), at least that is what seems to result from the interpretation of the limited data available on this subject.

Individual distributions can vary markedly from the average distribution, especially in cities and in industrial areas where the concentration of large nuclei is particularly high.

Measurements of atmospheric light scattering carried out by Fenn (1964) have shown that atmospheric nuclei in the $5 \times 10^{-2}\mu$ to 1μ radius range are divided into a discrete number of size groups, more or less well defined, separated by very pronounced minima of concentration. The number of groups increases with an increase in the total concentration of the nuclei, accordingly with an increase in pollution. The envelope of the concentration maxima follows Junge's size distribution law (Eq. 1), v decreasing from 3 in the case of a clear atmosphere to 2 in the case of an industrially polluted atmosphere.

With the aid of an aerosol photometer, Fenn, Gerber and Weickmann (1965) studied the size distribution of atmospheric nuclei in the range from 0.05μ to 1μ radius. The average distributions which they found in different areas free from atmospheric pollution are irregular and consist of several maxima separated by well defined minima. The curve of Junge (Eq. 1) gives a very good approximation of the envelope of the concentration maxima for the values of v varying with location, but in all cases falling between 2 and 4. One of these maxima, corresponding to a radius between 0.1μ and 0.15μ is observed at all continental stations and also, according to Goetz (1965) over the ocean. According to Fenn, Gerber and Weickmann (1965), this would seem to indicate that large nuclei of this size constitute the natural background of the atmospheric aerosol.

Large nuclei play an important role in: atmospheric chemistry where they represent more than 40% of the total mass of the nuclei (Junge, 1963b), in atmospheric optics because they scatter light and especially in the formation of atmospheric droplets (clouds, fog) because they constitute the veritable nuclei of condensation.

2.1.4. Giant nuclei

These nuclei can be studied by several methods: capture on plates or on rods exposed to the wind (see, for example, Woodcock, 1952),

capture by precipitation (see, for example, Junge, 1953), capture by sedimentation (see, for example, Junge, 1953), etc. As was the case with the large nuclei, the obtaining of samples presents no special difficulty, but their analysis is long and tedious.

According to Fig. 1 the average concentration of giant nuclei, the discovery of which is due to Woodcock and Gifford (1949), varies between 10 per cm^3 for $r = 1\mu$ and about 10^{-2} per cm^3 for $r = 10\mu$.

Over the ocean the concentration of this class of nuclei increases with wind speed, but the increase is more marked for the nuclei of great size (Woodcock, 1953).

The average concentration of giant nuclei also varies geographically. Isono, Komabayasi, Takahashi and Gonda (1966) estimated that for Japan this concentration falls between 10 and 10^2 nuclei per cm^3 . This order of magnitude is the same as that given earlier by Toba and Tanaka (1963) and is not very different from that found by Woodcock (1953) in the United States. In Hungary, according to Meszaros (1964), the concentration of giant chloride nuclei rarely exceeds 10^{-2} per cm^3 and on the average is on the order of 10^{-3} per cm^3 , a value which differs slightly from that found by Kumai (1965) in Alaska (0.9 to 1.6×10^{-3} per cm^3).

Variations in the concentration of giant nuclei have also been observed as a function of the atmospheric condition. As an example, according to Ramana Murty, Srivastava and Kapoor (1962) the average value of this concentration is significantly greater during the monsoon (July to September) than during the other months of the year. Although these authors have observed marked diurnal variations in the concentration of this class of nuclei, it can be said that the concentration is, in general, relatively constant, certainly more constant in all cases than with the large nuclei and especially than with the Aitken nuclei.

The giant nuclei are important in atmospheric chemistry; in effect they represent, in spite of their low concentration, more than 40% of the total mass of the nuclei (Junge, 1963b). These nuclei are of little effect in the formation of cloud droplets, but they seem, on the contrary, to play a fundamental role in the formation of rain.

2.1.5. Remarks

It follows from this brief examination of the granulometry of atmospheric nuclei that several methods may be simultaneously employed to obtain a complete spectrum of these nuclei. Considering the problems which such a determination poses (see, for example, Lodge, 1962),

it is not surprising that there are so few complete spectra of atmospheric nuclei.

Concerning the average spectrum of the continental aerosol (Fig. 1), it should be pointed out that it is of limited value, although based on a large number of observations, because these observations have been made by different methods, the accuracies of which are not necessarily the same.

Observations made by Metnieks (1958) in Ireland in maritime air masses have shown that chloride nuclei of radius greater than 0.15μ at 80% relative humidity pass through a maximum at a radius of about 0.25μ . This value is close to that found earlier by Rau (1955) and agrees relatively well with that estimated by Junge (1963b) in extrapolating the observations of Woodcock (1953). Recently, Goetz (1965) found that the distribution of the concentration of large nuclei exhibited a maximum for a radius between 0.15μ and 0.2μ when the ocean was calm and the wind speed from 0 to 5 km h^{-1} , which agrees with earlier results.

Taking these results into consideration, the differences in concentration of Aitken nuclei over the continent and over the ocean and of the composition of the atmospheric aerosol, Junge (1963b) estimated the most probable form of the average spectrum of an oceanic aerosol (Fig. 1, Curve 0). This spectrum has very limited value, especially since it is based on hypothesis for the part relative to Aitken nuclei and to large nuclei.

2.2. Granulometry aloft

2.2.1. Introduction

For a number of years it has been known that the atmospheric aerosol decreases with altitude up to the stratosphere.

Qualitatively this phenomenon is displayed in the upper troposphere and in the stratosphere in several ways: observations of nacreous clouds and crepuscular phenomena; observation of the decrease in atmospheric transparency from meteorite showers and from the color of the moon during eclipses of the moon; observation of dry haze layers from aircraft or from rising balloons; measurements of the electrical conductivity of the air, etc. (see, for example, Junge, Chagnon and Manson, 1961; Newkirk and Kroening, 1965).

However, quantitative observations of the stratospheric aerosol have only been available for a few years, thanks to the work of Junge, Chagnon and Manson (1961), of Chagnon and Junge (1961), of Junge and Manson (1961), of Junge (1961, 1963a), of Hodge (1961) and of Mossop (1965).

2.2.2. Aitken nuclei

As for the vertical variation of Aitken nuclei, the atmosphere may be divided into four layers (see Fig. 2).

CAPTION Figure 2. Mean vertical profiles of the concentration of Aitken nuclei as a function of altitude (Junge, 1963a)

1. Sioux Falls, United States
2. Hyderabad, India
3. Northeast United States (Weickmann, 1957)
4. Mean temperature profile, Hyderabad, India

The first layer extends from the ground up to an altitude of about 5 km. In this layer the decrease of the average concentration of these nuclei is exponential, the coefficients varying at first rapidly, then more slowly, with height. According to Junge (1961), the concentration of Aitken nuclei is from 60 to 600 nuclei per cm^3 at 5 km, the most frequent value being between 200 and 300 nuclei per cm^3 . These values are greater than those determined by Wigand (1919) and by Weickmann (1957) which, according to Junge (1961), resulted from the fact that these authors did not take into account the effect of the decrease in pressure with altitude in their measurements. They are less than those found by Selezneva (1965), which probably resulted from the fact that this author carried out his observations in summer, only when there was convection. The latter observations show important differences between the curves of the average values of concentration at different locations.

Individual curves of the vertical distribution of the concentration of Aitken nuclei in this layer differ markedly from the average curves to the point that one asks himself what this curve physically represents. The deviations are particularly important near the ground as confirmed by the observations carried out by Tester (1964) in the United States on the slopes of Little White Face Mountain between 365 and 1118 m altitude. Furthermore, it is a well-known fact that there are dry haze layers in the lower troposphere, layers which can be seen from the ground in an anticyclonic regime. Quite often one observes marked variations in the concentration of Aitken nuclei in passing through temperature inversions, as has been shown, for example, by Sagalyn and Faucher (1954), as well as Weickmann (1957). The concentration of these nuclei is also, in general, greater inside fair weather cumulus than outside as Weickmann (1966) has recently shown.

The distribution of concentration of Aitken nuclei in this portion of the atmosphere varies also according to season: the decrease with height is more marked in winter than in summer, which may be accounted for by the fact that convection is more intense during the warm portion

than during the cold portion of the year. This distribution also varies according to the general atmospheric situation. The decrease is certainly more marked in an anticyclonic regime than in a cyclonic regime; unfortunately, accurate data on this subject are lacking, the observations having been made nearly always in fair weather, i.e., anticyclonic regime. Also in this connection it is worthy of note that the average altitude of the top of this layer of the atmosphere is certainly greater than 5 km in cyclonic conditions.

The second layer of the atmosphere is one in which the average concentration of Aitken nuclei is quasi-constant. According to observations made at 44° north latitude in the United States, this layer extends from about 5 km to the tropopause, the average altitude of which was about 12 km at the time of the observations. The average concentration of the nuclei actually varies between 350 nuclei per cm^3 near 5 km and 200 nuclei per cm^3 at the tropopause, a variation which may be considered as practically nil compared to that observed between the ground and 5 km. The individual distributions, contrary to that which occurs in the lower troposphere, differ little from the average distribution. According to observations made at 17° north latitude over India, this quasi-constant average concentration of Aitken nuclei doesn't extend up to the tropopause, situated at about 17 km at the time of the observations, but up to about 13 km. This altitude coincides with a change of temperature gradient which, according to Junge (1963a), could correspond to an extension of the midlatitude tropopause beyond 30° north latitude toward tropical regions. In India the average concentration of Aitken nuclei varies between 200 nuclei per cm^3 near 5 km to 80 nuclei per cm^3 at 13 km, values less than those observed over the United States.

The third layer of the atmosphere, in which the average concentration of Aitken nuclei decreases exponentially, extends up to an altitude of about 17 km. It is located in the lower stratosphere over the United States and in the high troposphere over India. The decrease is more pronounced over the United States than over India; the average concentrations at 17 km are, respectively over these two areas of the globe, 1 nuclei and 10 nuclei per cm^3 .

The fourth layer of the atmosphere, in which the average concentration of Aitken nuclei is quasi-constant, extends up to about 27 km, the upper limit of reliable observations. The lower limit of detectability of the apparatus used is about 1 nuclei per cm^3 , an average value already reached at 17 km over the United States and at 20 km over India. In this connection, it should be noted that observations of nacreous clouds, which usually form between 25 and 30 km, indicate that the concentration of the aqueous particles constituting these clouds is several particles per cm^3 (see, for example, Hesstvedt, 1959).

There are no experimental data on the distribution of the concentration of Aitken nuclei as a function of their radius, in the stratosphere. Junge, Chagnon and Manson (1961) have theoretically estimated that the average radius of these nuclei was 0.04μ in hypothesizing that this distribution was the result of the combined action of the phenomena of coagulation and turbulent diffusion.

2.2.3. Large nuclei

For the vertical variation of large nuclei, the atmosphere may be divided vertically into three parts (see Fig. 3).

CAPTION Figure 3. Vertical profiles of the concentration of large nuclei as a function of altitude (Chagnon and Junge, 1961)

Individual stratospheric profiles in the northern part of the Middle west in the United States. Mean tropospheric profiles in Germany (Rossmann, 1950; Siedentopf, 1950). Mean tropospheric profile based on radiation measurements (Penndorf, 1954) and adjusted for agreement with the observations of Chagnon and Junge (1961) at 10 km altitude.

The first part, in which the average concentration of the nuclei decreases nearly exponentially (see Rossmann, 1950; Reeger and Siedentopf, 1950), extends from the ground up to an altitude of about 5 km. The second part, in which, according to qualitative optical measurements carried out by Penndorf (1954), this concentration is quasi-constant, extends from about 5 km to the tropopause. Combining these observations with those made near the tropopause over the United States where the altitude was about 12 km at the time of the measurements, Chagnon and Junge (1961) estimated that the boundary between these two parts of the atmosphere was situated at about 5 km and furthermore that the average concentration at this altitude was close to 3×10^{-2} nuclei per cm^3 .

The third part of the atmosphere, according to observations made over the United States, is characterized by the existence of a persistent layer of nuclei, situated between 16 and 23 km, in which the maximum concentration of approximately 10^{-1} nuclei per cm^3 is observed at about 20 km. Below this layer the average concentration of large nuclei increases almost exponentially from the tropopause where this concentration is, as we have seen, about 3×10^{-2} nuclei per cm^3 . Above this layer, the average concentration of large nuclei decreases up to about 27 km, the upper limit of the observations, where the concentration is variable and in the neighborhood, generally, of an average of 10^{-2} nuclei per cm^3 . Certain individual profiles seem to indicate a new increase in concentration of nuclei at 25 km; further observations are necessary before it can be decided if this increase is real or fictitious. The observations made in India, at 17° north latitude,

have shown a similar profile, the concentration maximum, however, being situated at a higher level but at practically the same distance from the tropopause. Aircraft observations at 20 km altitude have confirmed the existence of a continuous layer of large nuclei between the latitudes of 60°S and 70°N.

Similar profiles have been qualitatively shown by Elterman and Campbell (1964) from measurements of the light scattered from the beam of a vertically directed searchlight and by Bigg (1964) from measurements of the light scattered by the twilight sky. From simultaneous observations at different latitudes, this author has also shown that the vertical distribution of large nuclei retains the same form over horizontal distances not exceeding 300 km. It should also be noted that the altitude of the layers which strongly scatter light increases on the average from the pole to the equator.

There are few observations of the distribution of the concentration of large nuclei as a function of their radius in the troposphere. According to measurements of Fenn, Gerber and Weickmann (1965) at 2000m, 4000 m and 6000 m, this distribution is irregular and resembles that which these authors found near the ground.

There are more observations on the granulometry of large nuclei in the stratosphere. According to Junge, Chagnon and Manson (1961), the average distribution of concentration of these nuclei as a function of their radius can be represented by Eq. 1 in which $v = 2$. This straight line distribution differs from the parabolic distribution found by Mossop (1965) and by Friend (1966). (See Fig. 4.)

CAPTION Figure 4. Stratospheric distribution of the concentration n of large nuclei as a function of their radius. Curve 1, Chagnon and Junge, 1961; Curve 2, Mossop, 1965; Curve 3, Friend, 1966.

Further observations are necessary to establish whether these differences are real or due to the fact that the samples have not been taken on the same date or over the same locale or even if they appear and result from the method of reducing the data.

2.2.4. Giant nuclei

Current knowledge of the vertical distribution of giant nuclei in the lower troposphere is very contradictory; therefore, we shall restrict ourselves to extricating the broad outline.

Above the oceans the average concentration of giant sea salt nuclei in general decreases almost exponentially with altitude. In spite of this decrease the distribution of the concentration as a function of the

dimension of the nuclei remains almost constant. Occasionally, especially when the wind is light, the concentration increases for a few hundred meters before decreasing (see Lodge, 1955; Woodcock, 1952 and 1957; Durbin and White, 1961).

Above the continents, according to Byers, Sievers and Tufts (1957), the average concentration of giant sea salt nuclei is relatively constant with altitude except for several tens of meters close to the ground where it decreases sharply. These authors attribute this reduction to the capture of the nuclei by obstacles situated near the ground such as trees. In Czechoslovakia, Podzimek and Cernoch (1961), in disagreement with Byers, Sievers and Tufts (1957), have measured the greatest concentrations near the ground and have concluded, on that basis, that the majority of giant chloride nuclei was of continental origin. In Hungary, Meszaros (1964) found profiles in agreement with those of these authors, the decrease, however, ceasing at several hundreds of meters above the ground, which he attributed principally to thermal convection.

These contradictions are probably only apparent, the vertical distribution of the giant nuclei depending strongly on the geographical and thermal conditions. According to Laktionov (1960), the concentration of nuclei of radius greater than 4μ is relatively constant under halcyon skies between 100 and 1000 m. The value of this concentration is, however, strongly dependent on the nature of the ground cover; it is, for example, 100 times lower over snow and over the ocean than over the desert and the steppes. Also, according to Semonin (1966) certain industrial zones are very good sources of giant hygroscopic nuclei, which brings up the question of knowing up to what point the hypothesis that giant chloride nuclei observed at altitude far in the interior of the continent (Crozier, Seely and Wheeler, 1952; Twomey, 1955; Byers, Sievers and Tufts, 1957) are of maritime origin is acceptable. Other systematic observations are necessary to clarify the answers to these questions.

Limited data available on the granulometry of giant nuclei in the upper troposphere and in the stratosphere indicate that they are not numerous which, based on their mass, appears reasonable. According to Junge, Chagnon and Manson (1961) it seems that their concentration varies markedly with time in the stratosphere.

2.2.5. Remarks

The fact that the profile of Aitken nuclei in the troposphere is qualitatively the same as that of the large nuclei proves that the effect of sedimentation is not involved in the establishment of these profiles, since they are independent of the radius of the nuclei. This result is theoretically explicable since calculation indicates that sedimentation begins to be effective only for those giant nuclei of great size.

It should be noted that the concentration of Aitken nuclei in the upper troposphere is quasi-constant over the continents and of the same order of magnitude as at the surface of the ocean. This concentration of 100 to 300 nuclei per cm^3 is close to the concentration of 200 nuclei per cm^3 observed by Dav (1955) in pure maritime air at 2 km altitude near the coast over England and the Atlantic. These facts led Junge (1963b) to acknowledge that the Aitken nuclei concentration is nearly constant in the troposphere both horizontally and vertically, except for the first five kilometers over the continents and in their proximity over the ocean. Therefore, the continents constitute the source of Aitken nuclei which spread out vertically by turbulent diffusion and horizontally by advection. As for the average concentration of 200 to 300 nuclei observed in the upper troposphere over the continents and all the troposphere over the oceans, it constitutes an equilibrium value resulting from the action of different physical processes acting on this concentration.

The preceding considerations also theoretically apply to large nuclei. Unfortunately there is very little observational data on this class of nuclei on which to verify this hypothesis.

The decrease of the concentration of Aitken nuclei above the tropopause implies that the majority of these nuclei are of tropospheric origin. Different processes can be envisioned to explain how they penetrate into the stratosphere; actually there are too few data upon which to base a theory on this subject.

According to Junge, Chagnon and Manson (1961), the shape of the profile of the nuclei in the stratosphere indicates that these nuclei ought to form in situ between 16 and 23 km (see 3.1.3.). The existence of a continuous layer of large nuclei in this part of the atmosphere can in effect be difficult to explain otherwise, unless the nuclei penetrate and spread out in the stratosphere, which does not seem probable. It is not the opinion of Martell (1966) who, based on observations of the radioactivity of atmospheric nuclei and on chemical considerations relative to their composition, thinks that there are many sulfate Aitken nuclei of great size in the upper troposphere which could explain the formation of large nuclei in the stratosphere by coagulation

3. Chemical properties of the nuclei

3.1. Formation of the nuclei

3.1.1. Classification of the mechanisms of formation

There are a great number of mechanisms for forming atmospheric nuclei, mechanisms which may be classified in four categories according

to whether their formation results from combustion, chemical reactions in the atmosphere, the dislodging of liquid or solid particles from the surface of the earth or the entry of particles of cosmic origin into the atmosphere.

3.1.2. Mechanisms resulting from combustion

At the time of natural (volcanic eruptions, brush fires, etc.) or artificial (domestic fires, industrial fires, etc.) combustion, the volatile products of combustion evaporate or sublime and emerge into the atmosphere at an elevated temperature. The vapors thus formed, of which the saturation pressure is in general low, cool quickly by mixing with the atmospheric air and because of this become highly supersaturated (a few hundreds and even thousands of percent). As a consequence of this high supersaturation, there are formed, by homogeneous or heterogeneous nucleation, nuclei of which the radii are inversely dependent on the degree of supersaturation; in general these are Aitken nuclei but also large nuclei and even giant nuclei.

The nonvolatile combustion products which emerge into the atmosphere are burned or partially burned products. They are, consequently, friable and break easily under the action of the wind. The nuclei thus formed have an average radius of the order of 1μ and are, therefore, mostly large nuclei.

Principally in the troposphere and especially in the lower troposphere these mechanisms are active. They can also cause the formation of nuclei in the stratosphere, in particular at the time of volcanic eruptions, as shown by Mossop (1964).

3.1.3. Mechanisms resulting from chemical reactions

In the atmosphere there is a wide variety of gases of low concentration. They are formed principally in industrial regions during combustion. They also result from the decomposition of animal and plant substances, from animal and plant life, and from the fermentation in swamps, etc.

Principally under the action of heat, of humidity, or of short wavelength radiation, certain of these gases can inter-react. For example, NH_4Cl can be formed by the reaction of NH_3 and of HCl , or SO_3 can be oxidized to form H_2SO_4 ; still other reactions are possible. The qualitative study of these reactions has been tackled, but their quantitative study remains to be done (see, for example, Cauer, 1951 and Junge, 1964b). In general the nuclei which are formed are also Aitken nuclei.

The substances resulting from these reactions can also give birth to large nuclei or to giant nuclei. According to Junge and Manson (1961), that is what would be produced in the stratosphere where the sulfate, resulting from the oxidation of SO_2 or of H_2S under the effect of ozone and short wavelength radiation, would agglomerate around Aitken nuclei to form large nuclei.

3.1.4. Mechanisms resulting from the release of particles from the surface of the earth

3.1.4.1. Release of liquid particles

Two mechanisms are usually called upon to explain the formation of nuclei on the oceans: atomization of water thrust upward from the sea (spray) under the action of the wind and bursting of air bubbles at the surface of the sea.

The sprays in general consist of large droplets which, by virtue of their fall speed, cannot remain in suspension in the air for very long and therefore don't penetrate deeply into the interior of the land.

Facy (1951) pointed out that the assemblage of small waves forming foam give birth, outside of a few large droplets comparable in size to those of fog, to a great number of very small droplets of radius between 0.02μ and 1μ . The same phenomenon is produced at the time of the bursting of bubbles which form from air, entrapped by the falling back of the crest of the wave, reappearing at the surface of the water. According to Blanchard and Woodcock (1957), the radius of these bubbles is quite variable, but less than 100μ for the majority of them.

The mechanism of bursting of bubbles has been studied using high speed photography. Kientzler, Arons, Blanchard and Woodcock (1954) as well as Knelman, Dombrowski and Newitt (1954), found that the bubbles became thinner in the upper portion as they reached the surface, then burst; at the time of bursting, the water which falls in the cavity which is formed emerges at the center of this depression in an unstable vertical jet the upper part of which breaks, giving rise to several large droplets (see Fig. 5). The relation between the radius of these droplets and that of the bubble which gave rise to them has been examined by Moore and Mason (1954) who found that the radius of the droplets is about ten times smaller than that of the bubble which gave rise to them.

CAPTION Figure 5. Mechanism of release of bubbles from the surface of the sea (Mason, 1954). The large droplets, G, are formed when the vertical jet breaks and the small droplets, g, when the upper part of the bubble bursts.

The mechanism of the formation of fine droplets at the time of the bursting of the upper part of the bubble is difficult to study because the duration of the bursting is on the order of a microsecond and also because these droplets are so small that they evaporate rapidly at normal humidity. The first quantitative observations of the number of these droplets are due to Mason (1954, 1957). He showed that the rupture of the wall of a large bubble of sea water produces hundreds of droplets, whatever the radius of the bubble, a result which has been confirmed by Isono (1959) and Twomey (1960). Blanchard (1963) (see Day, 1964) found, on the contrary, that the number of small droplets thus formed is not constant and that it is a function of the radius of the bubble; a bubble of 0.15 mm radius produces two droplets and a bubble of 3 mm radius, about 1000 droplets, at the maximum. Recent results by Day (1964) agree relatively well with those of Blanchard but differ, however, from those of Mason. In effect Day found that the average number of small droplets which form at the time of the bursting of a bubble of salt water varies from 0, when the radius of the bubble is 0.05 mm (50 μ), to between 300 and 400, when the radius is 2 mm. However, it should be noted that the results obtained by these three authors agree for large bubbles ($r > 0.5$ mm).

Day (1964) also showed the influence of salinity on the number of droplets in the sense that the droplets are definitely more numerous in the case of salt water than distilled water.

Given the low concentration of salt in sea water, the saturation pressure of droplets formed by the mechanisms set forth differs little from the saturation pressure of droplets of pure water. Since the humidity is generally greater over the oceans, only the small droplets will be sufficiently displaced from the surface by turbulent diffusion to reach the layers of air in which they evaporate. If the evaporation is complete, the sea salt which they contain crystallizes; during the course of their existence, these small droplets can pass from the liquid state to the solid state several times.

Dessens (1946, 1949) has shown that the salt contained in the droplets shatters when it crystallizes at low relative humidities. A quantitative study of this process carried out by Lodge and Baer (1954), did not reveal a measurable increase in the concentration of nuclei; that results from the fragmentation of large crystals but even this is not always the case. Blanchard and Spencer (1964) arrived at the same conclusion in studying the crystallization of droplets, the radius of which varied between 2.5 μ and 50 μ , droplets composed of sea water, of concentrated sea water, or of an aqueous solution containing 3.5% of sodium chloride. Indeed these authors have never noted the formation of small salt nuclei at the time of droplet crystallization. Twomey and McMaster (1955), on the contrary, observed that large droplets of a sea salt

solution give birth, when they crystallize, not only to a large particle of salt, but also to a large number of very small fragments. Opinions are thus divided on this question; however, it seems unlikely that sea salt disintegrates when it crystallizes, and the small nuclei composed of this substance ought, therefore, to have a different origin.

3.1.4.2. Release of solid particles

A large part of the continent is covered with solid particles (sand, dust, etc.) which, under the action of the wind, are released from the ground and dispersed in the atmosphere. This source of nuclei is of relatively little importance in regions of high rainfall; but this is not the case for arid regions where the finest of these particles can be lifted very high and, because of this, be transported great distances. On the other hand, the large particles often disintegrate under the mechanical action of the wind giving rise to small particles which can spread through all the troposphere.

In general, the particles released from the soil are of large nuclei or of giant nuclei. Except for the case of violent sand or dust storms, the giant nuclei fall rapidly to the ground and their zone of dispersion is thus limited to the vicinity of their source.

3.1.5. Mechanisms resulting from the entry into the atmosphere of particles of cosmic origin

It is certain that particles of cosmic origin burn or melt when they enter the atmosphere giving rise to nuclei which, for the most part, are probably Aitken nuclei. There are very few direct observations of nuclei thus formed but their concentration ought to be normally low in the lower stratosphere.

In this connection it should be pointed out that the large sulfate nuclei observed in the stratosphere (see 3.1.3.) contain fine, solid, insoluble particles the modal radius of which is about 0.04μ , nuclei similar to those captured at 80 km by rockets (Hemenway, Soberman and Witt, 1963).

Bowen's hypothesis (1953, 1956), according to which certain periodic variations of precipitation result from an influx of ice nuclei formed in the atmosphere at times of meteoritic showers, has attracted attention to nuclei of cosmic origin. This hypothesis has already caused much ink to flow and will continue to foster much, such that the research of this subject will be even more effective than today not only in verifying this hypothesis but in confirming it and at times in refuting it. No further attention will be focused on this question because of the conviction that the portion of the atmospheric aerosol which is of cosmic origin plays only a secondary role in meteorology.

3.2. Chemical composition of the atmospheric aerosol

Following the last world war a new, or rather little previously developed, discipline became established: atmospheric chemistry. Concerning the atmospheric aerosol, one actually has a general idea of its composition "en masse". On the contrary, in spite of the development of microchemical techniques, there exist only fragmentary data on the chemical composition of individual nuclei, because of the difficulty in applying these techniques to such small particles as nuclei (see, for example, Junge, 1963b).

In this paper we shall limit ourselves to a few words on the composition of condensation nuclei, about which the atmospheric droplets form and of ice nuclei which cause or favor the formation of atmospheric ice crystals.

The large nuclei constitute the true nuclei of condensation. Giant nuclei also play this role but, because of their low concentration, are insignificant and can be neglected. As for Aitken nuclei, they become significant only in regions where the number of large nuclei is insufficient, which is probably the case over the oceans.

The majority of the condensation nuclei are mixed nuclei (Junge, 1952) composed of a solid, insoluble particle surrounded by soluble, hygroscopic substances. Microphotography of the evaporative residue of cloud droplets shows, as a matter of fact, that most of the nuclei were apparently droplets before evaporation of the liquid part in the field of the electron microscope. That has also been indirectly demonstrated in simultaneously collecting the atmospheric aerosol at various humidities on clean, dry plates, which capture only droplets, and on plates covered with a film of a viscous substance which captures both droplets and dry particles. Also it may be noted that a considerable portion of atmospheric nuclei act as if they were dry when the relative humidity is less than 70% and as if they were droplets when the relative humidity is greater than this value.

From a certain relative humidity, generally between 70 and 80%, upward the condensation nuclei consist of solution droplets often containing a solid, insoluble particle. Authors persist in terming these droplets condensation nuclei, which sometimes leads to confusion. This is why a distinction should be made between condensation nuclei which, in principle, are solid but can be deliquescent and droplet-nuclei which are liquid and are already in fact atmospheric droplets.

Condensation nuclei have been studied individually with an electron microscope by Junge (1953), Kuroiwa (1953, 1956), Yamamoto and Ohtake (1955), Isono (1959), and Kumai (1965). This method is interesting, but it permits the identification of only the crystalline constituents and it gives no indication of their relative concentration. These observations are evidence that the condensation nuclei are for the most part composed of sea salt,

combustion products and particles released from the ground. The percentage of these three types of nuclei differs according to author; those of sea salt nuclei, for example, vary between 10 and 90%. With the available data it is difficult at this time to explain the origin of these variations.

When crystals of natural ice are sublimed, in general one large particle is found in the central part of the crystal and a variable number of very small particles throughout the crystal. Most authors admit that this large particle is the ice nuclei around which the crystal forms. Several authors (see, for example, Mossop, 1963 and Vali, 1966) have remarked that there is no preemptory reason for assuming that this large particle, rather than one of the smaller ones, gives rise to the formation of the crystal. It is difficult to settle this question with the available data; pending proof to the contrary we shall assume that the large particle is the true ice nucleus.

Individual ice nuclei have been studied by electron microscopy by Kumai (1951, 1957, 1961), aufm Kampe, Weickmann and Kedesdy (1952), Isono (1955, 1959), Kumai and Francis (1962), Kumai and O'Brien (1965) and Rucklidge (1965). These observations have shown that these nuclei mainly consist of mineral particles; from 57 to 88% in nonpolluted regions. Measurements of the threshold activity of samples of different soils have indirectly confirmed these results. Indeed these observations show that this threshold of activity varies between -5°C and -17°C , temperatures between which the first ice crystals appear in natural clouds.

According to Bigg and Miles (1964), soil particles do not constitute the majority of ice nuclei in Australia, which seems to confirm the determinations by Paterson and Spillane (1967) of the threshold of activity of particles of several soils from the arid Australian zone. Ice nuclei in this region of the earth could thus be mainly of cosmic origin, which would confirm Bowen's hypothesis (see 3.1.5.).

4. Physical properties of nuclei

4.1. Movement of the nuclei

4.1.1. Resistance of the air to movement of the nuclei

Air presents a resistance to the movement of nuclei, the action of which is that of a force F which acts in a direction opposite to that of the movement. The form of this force depends on the relation which exists between the radius r of the nucleus (assumed to be spherical) and the mean

free path l of the molecules of air, free path approximately equal to 0.1μ in normal conditions of temperature and of pressure at the earth's surface.

When the radius of the nucleus is very small in relation to the mean free path of the molecules ($r \ll l$), i.e., when the movement of the nucleus does not perturb the law of the distribution of speed of the molecules (see, for example, Fuchs, 1964),

$$F = \frac{6\pi n r^2 v}{Kl} \quad (3)$$

where v is the speed of the nucleus, n the coefficient of dynamic viscosity of the air, K is a numerical coefficient the value of which depends on the hypothesis made concerning the nature of the impacts between the molecules and the nucleus, a coefficient which can, as a first approximation, be assumed to be 1.

When the radius of the nucleus is very large in relation to the mean free path of the molecules ($r \gg l$), i.e., when the movement of the nucleus perturbs the molecular distribution law and gives rise to hydrodynamic flows, the force F is given by Stokes' Equation (see, for example, Fuchs, 1964).

$$F = 6\pi n r v \quad (4)$$

In order to establish this formula, it is assumed that there does not exist a discontinuity in speed near the surface of the nucleus, i.e., there exists a thin film of air adjacent to the surface and stationary with respect to the nucleus. This assumption is no longer valid when the radius of the nucleus is not very large with respect to the mean free path of the molecules. In order to take into account the gradient of speed which exists in the air near the surface of the nucleus under that condition, it is necessary to introduce a correction factor into the Stokes Equation which then becomes the Cunningham Equation (see, for example, Fuchs, 1964)

$$F = \frac{6\pi n r v}{1 + A\left(\frac{l}{r}\right)} \quad (5)$$

where A is a numerical coefficient, the value of which depends on the nature of the surface of the nucleus and which can, as a first approximation, be set equal to 2.

Semiempirical formulas giving the value of F have been proposed when the radius of the nucleus is neither very small nor very large in relation to the free path of the molecules. In comparing values of F derived from

these formulas with values of F drawn from formulae (3), (4), and (5), Fuchs (1964) established the following table:

TABLE 3

Limits of applicability of several formulae for calculating the air resistance to the movement of nuclei

Formula	Admissible Error	
	1%	10%
(3)	$1 \times 10^{-3} \mu < r < 2 \times 10^{-3} \mu$	$5 \times 10^{-4} \mu < r < 2 \times 10^{-2} \mu$
(4)	$8 \mu < r < 15 \mu$	$8 \times 10^{-1} \mu < r < 35 \mu$
(5)	$1.8 \times 10^{-1} \mu < r < 8 \times 10^{-1} \mu$	$5 \times 10^{-2} \mu < r < 8 \mu$

From this table, if the admissible error is only 1%, the interval of applicability of each of these formulae is narrow and the ensemble of these intervals does not cover the gamut of atmospheric nuclei radii. In contrast, if an error of 10% is acceptable, formula (4) of Stokes is completely suitable for giant nuclei and formula (3) for Aitken nuclei, except for the largest of these nuclei. As for formula (5) of Cunningham, it is simultaneously suitable for the large Aitken nuclei, the large nuclei and the smallest giant nuclei.

4.1.2. Terminal fall speed of the nuclei

Consider a spherical nucleus of radius r and of density ρ which falls at a speed v in calm air of density ρ' and dynamic viscosity coefficient η .

Assume the nucleus initially at rest at time $t = 0$. If O_z is the vertical axis through the initial position O which is taken as the origin of the O_z axis, the differential equation of motion may be written:

$$\frac{d^2 z}{dt^2} = \frac{dv}{dt} = g - \frac{F}{m} \quad (6)$$

where g is the acceleration due to gravity and where

$$m = \frac{4}{3} \pi r^3 (\rho - \rho') \quad (7)$$

When the nucleus reaches its terminal speed ($v = C_{ste}$), Equation (6) may be written:

$$mg = F \quad (8)$$

From (4), (5), (7), and (8), the terminal speed of the nuclei is given by the relation:

$$v_1 = \frac{2g(\rho - \rho')r^2}{9\eta} \quad (9)$$

for the nuclei for which Stokes Formula (4) is applicable and by the formula

$$v_1 = \frac{2g(\rho - \rho')r^2}{9\eta} \left(1 + A\frac{1}{r}\right) \quad (10)$$

for the nuclei for which the Cunningham Formula (5) is applicable.

Assume that the atmospheric pressure is equal to 1 atmosphere (1013.25 mb), the temperature 293°K, the relative humidity 50% and the acceleration of gravity 980 cm sec⁻², in which case $\rho' = 1.20 \times 10^{-3}$ g cm⁻³ and $\eta = 1.818 \times 10^{-4}$ g cm⁻¹ sec⁻¹. The densities of the nuclei are not well known. Apparently they ought to be between 1 and 2 g cm⁻³. For $\rho = 2$ g cm⁻³, formulae (9) and (10) give the values listed in Table 4 for the terminal fall speed of the nuclei.

TABLE 4

Terminal fall speed of a nucleus as a function of its radius.

Stokes Formula (9)		Cunningham Formula (10)	
$r(\mu)$	$v_1(\text{cm/s})$	$r(\mu)$	$v_1(\text{cm/s})$
40	38.31	5	6.29×10^{-1}
20	9.58	1	2.63×10^{-2}
10	2.39	5×10^{-1}	7.18×10^{-3}
5	5.99×10^{-1}	1×10^{-1}	4.79×10^{-4}
1	2.39×10^{-2}	5×10^{-2}	17.96×10^{-5}

From this table it is seen that, except for the largest of the giant nuclei, the terminal fall speed is very low; it can be agreed that it is negligible when the radius of the nucleus is less than 1 μ .

4.1.3. Effect of turbulence on the sedimentation of nuclei

Let n be the concentration of the nuclei and the accumulation of nuclei $\frac{\partial n}{\partial t}$ in a unit volume in a unit time be equal to the convergence (divergence with a change of sign) of the flux $\vec{\phi}$ of the nuclei in this volume

$$\frac{\partial n}{\partial t} = -\text{div} \vec{\phi} \quad (11)$$

In the case under consideration this flux is composed of a diffusive turbulent flux $-D\nabla n$ where D is the coefficient of turbulent diffusion of the nuclei and ∇n the ascendant of n , as well as an advective flux $n\vec{v}$ where v is the speed of the nuclei. Equation (11) may then be written

$$\frac{\partial n}{\partial t} = -\text{div}(-D\nabla n + n\vec{v}) \quad (12)$$

By placing the z axis along the vertical descendant and by assuming homogeneity in the horizontal plane, (12) then takes the following form:

$$\frac{\partial n}{\partial t} = \frac{\partial}{\partial z} (D \frac{\partial n}{\partial z} + v_1 n) \quad (13)$$

where the nucleus speed v has been replaced by its terminal speed v_1 .

Assuming that the regime is permanent ($\frac{\partial n}{\partial t} = 0$) and integrating Equation (13) with the limiting condition that $n = 0$ for $z = \infty$,

$$D \frac{dn}{dz} + v_1 n = 0 \quad (14)$$

The coefficient of turbulent diffusion D is a function of the altitude z which, over the ocean, can be expressed by the following formula according to Junge (1957):

$$D = \frac{A_\infty u_5 k(z+b)}{A_\infty + \rho' u_5 k(z+b)} \quad (15)$$

where A_∞ is the exchange coefficient for $z \rightarrow \infty$, u_5 the wind speed at a height of 5 m, k Karman's constant, b the roughness parameter and ρ' the density of air.

Integrating Equation (14), taking (15) into account, with the condition that $n = n_0$ for $z = 0$, (see Junge, 1957)

$$\frac{1}{\epsilon} = \frac{n_0}{n} = \left(\frac{z+b}{b}\right)^{\frac{v_1}{u_5 k}} \exp\left(\frac{\rho' v_1 z}{A}\right) \quad (16)$$

where ϵ is the ratio of the nuclei concentration at altitude z to that at the ground ($z = 0$).

From Tables 5a and b may be found the values of the ratio ϵ as a function of the radius r of the nuclei and of the height z above the ocean for $k = 0.4$, $b = 4$ cm, $\rho = 2 \text{ g cm}^{-3}$, $\rho' = 1.2 \times 10^{-3} \text{ g cm}^{-3}$ and for the value of v_1 calculated from formula (9).

TABLE 5

Value of the ratio ϵ as a function of the radius r of the nuclei and of the height z above the ocean

a. $A = 100 \text{ g/cm, s; } u_5 = 500 \text{ cm/s}$

$r(\mu)$	$z(\text{m})$ 10	50	100	200	300	400
1	0.9991	0.9977	0.9962	0.9933	0.9904	0.9875
5	0.977	0.944	0.909	0.844	0.785	0.730
10	0.909	0.795	0.683	0.508	0.380	0.284
15	0.808	0.597	0.424	0.218	0.113	0.059
20	0.684	0.400	0.217	0.067	0.021	0.006
25	0.553	0.239	0.092	0.015	0.002	0.000
30	0.426	0.127	0.032	0.003	0.000	0.000

b. $A = 400 \text{ g/cm, s; } u_5 = 2,000 \text{ cm/s}$

$r(\mu)$	$z(\text{m})$ 10	50	100	200	300	400
1	0.9998	0.9994	0.9990	0.9983	0.9976	0.9969
5	0.994	0.986	0.976	0.959	0.941	0.924
10	0.977	0.944	0.909	0.844	0.785	0.730
15	0.948	0.879	0.807	0.683	0.580	0.492
20	0.909	0.795	0.683	0.508	0.380	0.284
25	0.862	0.699	0.551	0.347	0.220	0.140
30	0.808	0.597	0.424	0.218	0.113	0.021

Examination of these two tables reveals that the value of ϵ decreases with height and more rapidly than the radius of the nuclei increases. This decrease is very small for nuclei of radius less than 1μ , i.e., for large nuclei and Aitken nuclei. Also in comparing these two tables

is seen that, for a given radius, the decrease of ϵ with height is weaker the stronger the turbulence.

If, as a first approximation, it is assumed with Junge (1957) that the effects of turbulent diffusion and of sedimentation are compensated when $\epsilon < 0.2$, it appears that according to whether the turbulence is weak (Table 5a) or strong (Table 5b) the nuclei of, respectively, 20 μ and 30 μ are hardly lifted above the ground.

Above the continents, because of the complexity of the phenomenon due mainly to the irregularity of the surface of the ground and also to the fact that in industrial regions the nuclei are introduced at a certain height (chimneys), it is difficult to establish a formula analogous to (15). According to Junge (1957), it seems that the general trend of the phenomenon is the same over the continents as over the oceans, the upper limit of penetration being somewhat greater in the first case than in the second.

4.2. Coagulation of the atmospheric aerosol

4.2.1. General

By coagulation of an aerosol is meant the process by which the constitutive particles of this aerosol adhere to one another when they collide, a process which in effect reduces their concentration. All collisions between particles do not necessarily result in an adherence; practically, it seems that this is a good hypothesis in the case of an atmospheric aerosol consisting of mostly liquid particles.

Several phenomena can cause the coagulation of aerosols. In the case of the atmospheric aerosol, it can result: from differences in the fall speeds of the nuclei (gravitational coagulation), from the presence of an electric field and of charges which the nuclei can carry (electrical coagulation), from turbulence (turbulent coagulation) and finally from Brownian motion (thermal coagulation).

Because of the slow fall rate of nuclei (see 4.1.2.), gravitational coagulation is significant only for giant nuclei for which the concentration, in comparison to the other nuclei, is so low (see 2.1.4.) that this form of atmospheric coagulation may be neglected. Considering the charges which the nuclei normally carry and the intensity of the electric field which normally exists in the atmosphere, electrical coagulation can, as well, in general be neglected (see, for example, Fuchs, 1964). Turbulent coagulation can also be neglected (see, for example, Fuchs, 1964). Such, however, is not the case with thermal coagulation which, as shall be seen, plays an important role in the establishment of the granulometry of the atmospheric aerosol.

4.2.2. Fundamental equations of Brownian motion

Brownian motion refers to the irregular, thermally induced motion of small particles suspended in a fluid.

The fundamental equations of Brownian motion are the following (see, for example, Duclaux, 1938 and Fuchs, 1964):

$$\overline{x^2} = 2 Dt \quad (17)$$

$$D = kTB \quad (18)$$

where $\overline{x^2}$ is mean square of the displacement of the particles along the x-axis during time t, D the coefficient of molecular diffusion of the particles, k the Boltzmann constant, T the absolute temperature and B the mobility coefficient of the particles.

Recall that, by definition, the mobility coefficient is given by the relation

$$B = \frac{v}{F} \quad (19)$$

where v is the speed of the particle and F is the resistance of the air to its movement. From the speed as given by formula (3) or by formula (5) it then follows that

$$B = \frac{Kl}{6\pi\eta r^2} \quad (20')$$

$$B = \frac{1 + A(\frac{1}{r})}{6\pi\eta r} \quad (20'')$$

If the aerosol is isotropic, i.e., if the diffusion coefficient of the particles is independent of the direction of their movement, it may be deduced from (17) (see, for example, Fuchs, 1964) that

$$\frac{\partial n}{\partial t} = D \operatorname{div}(\nabla n) = D \nabla^2 n \quad (21)$$

where n is the concentration of the particles, ∇n the ascendant and $\nabla^2 n$ the Laplacian of this concentration.

4.2.3. Capture of nuclei by a sphere

Consider a stationary sphere of radius R in an unbounded aerosol. The aerosol being assumed isotropic, (21) may be written

$$\frac{\partial n}{\partial t} = D \left(\frac{\partial^2 n}{\partial r^2} + \frac{2}{r} \frac{\partial n}{\partial r} \right) \quad (22)$$

where n is the nuclei concentration at time t at distance r ($r > R$) from the center of the sphere under consideration.

By assuming that the regime is permanent ($\frac{\partial n}{\partial t} = 0$), (22) may be written

$$\frac{\partial^2 n}{\partial r^2} + \frac{2}{r} \frac{\partial n}{\partial r} = 0 \quad (23)$$

Integrating this equation under the following limiting conditions

$$n = n_0 \text{ for } r = \infty \text{ and } n = 0 \text{ for } r = R, \quad (24)$$

the latter condition expressing that each collision of a nucleus with the sphere results in capture, yields

$$\frac{\partial n}{\partial r} = - \frac{R n_0}{r^2} \quad (25)$$

Let N represent the number of nuclei which collide with the sphere. From (21) it follows that

$$\frac{\partial N}{\partial t} = \iiint_V D \operatorname{div}(\nabla n) \delta v = \iint_{\text{sphere } R} D (\nabla_N n) \delta \sigma \quad (26)$$

where $\nabla_N n$ is composed of the ascendant ∇n along the normal exterior to the sphere.

Because of the spherical symmetry, (26) may be simplified and becomes

$$\frac{\partial N}{\partial t} = \iint_{\text{sphere } R} D \left(\frac{\partial n}{\partial r} \right)_{r=R} \delta \sigma = 4 \cdot R^2 D \left(\frac{\partial n}{\partial r} \right)_{r=R} \quad (27)$$

or also, as calculated from (25),

$$J = \frac{\partial N}{\partial t} = 4\pi R D n_0 \quad (28)$$

where J is the rate of capture of nuclei by the sphere due to Brownian motion.

4.2.4. Thermal coagulation of the atmospheric aerosol

Consider a spherical nucleus of radius r_1 , assumed stationary, which, as a result of Brownian motion, captures nuclei of radius r_2 . Since the spherical nuclei enter contact when the distance of their centers is equal to the sum of their radii, the assumed stationary particle may be replaced by a "captive sphere" of radius $r_1 + r_2$ and the other particles by their center.

Let us drop the hypothesis of the immobility of the "captive sphere". In Equation (17), the mean square $\overline{x^2}$ of the displacement of the particle should then be replaced by the mean square $\overline{(x_1 - x_2)^2}$ of the relative displacement of the nucleus of radius r_2 in relation to the one of radius r_1 and thus

$$\overline{(x_1 - x_2)^2} = 2D_{12}t \quad (29)$$

where D_{12} is the relative diffusion coefficient of the two nuclei.

But

$$\overline{(x_1 - x_2)^2} = \overline{x_1^2} + \overline{x_2^2} - 2\overline{x_1 x_2}$$

or, even further, Equation (17) having been established in admitting the independence of the displacements of the particles,

$$\overline{(x_1 - x_2)^2} = \overline{x_1^2} + \overline{x_2^2} \quad (30)$$

From (17) and (29) it follows that

$$D_{12} = D_1 + D_2 \quad (31)$$

where D_1 and D_2 are, respectively, the diffusion coefficients of particles of radii r_1 and r_2 .

For the case under consideration, Equation (28) may be written

$$J_2 = 4\pi(r_1 + r_2)(D_1 + D_2)n_2 \quad (32)$$

where J_2 is the rate of capture of nuclei of radius r_2 by a nucleus of radius r_1 . If all nuclei have the same radius ($r_1 = r_2 = r$), formula (32) may be written ($D_1 = D_2 = D$)

$$J = 16\pi r D n \quad (33)$$

where D is the diffusion coefficient of nuclei of radius r and n the concentration of these nuclei.

If n is the concentration of nuclei of radius r_1 , the rate of capture of nuclei of radius r_2 by all the nuclei of radius r_1 per unit volume is given by the formula

$$J_{1,2} = 4\pi(r_1 + r_2)(D_1 + D_2)n_1n_2 \quad (34)$$

Each time a nucleus of radius r_1 collides with a nucleus of radius r_2 , the total number of nuclei is reduced by two units, but since a nucleus of radius $r_1 + r_2$ is formed, the volume concentration $n = n_1 + n_2$ of the nuclei in reality is reduced by only one unit. Therefore,

$$\frac{\partial n}{\partial t} = -4\pi(r_1 + r_2)(D_1 + D_2)n_1n_2 \quad (35)$$

If all the nuclei have the same radius ($r_1 = r_2 = r$), formula (35) takes the following form

$$\frac{\partial n}{\partial t} = -16\pi r D n^2 \quad (36)$$

If in (35) D_1 and D_2 are replaced by their values taken from (18) it follows that

$$\frac{\partial n}{\partial t} = -4\pi kT(B_1 + B_2)(r_1 + r_2)n_1n_2 \quad (37)$$

Consider first the case where formula (5) is applicable; from (20") it follows then that

$$\frac{dn}{dt} = -\frac{2}{3} \frac{kT}{r} \left[\frac{1}{r_1} + \frac{1}{r_2} + A\ell \left(\frac{1}{r_1^2} + \frac{1}{r_2^2} \right) \right] (r_1 + r_2) n_1 n_2 \quad (38)$$

If $r_1 \gg r_2$ and if $n \approx n_2 \gg n_1$, formula (38) takes the following form

$$\frac{dn}{dt} \approx -\frac{2}{3} \frac{kT}{r} \left(1 + \frac{A\ell}{r_2} \right) \frac{r_1}{r_2} n_1 \quad (39)$$

By integrating this equation between the initial state (t_0, n_0) and some state (t, n) ,

$$\ln \frac{n}{n_0} \approx -\frac{2}{3} \frac{kT}{r} \frac{r_1}{r_2} \left(1 + \frac{A\ell}{r_2} \right) n_1 (t - t_0) \quad (40)$$

If the half life, i.e., the time $(t - t_0)$ necessary for $\left(\frac{n}{n_0}\right) = 0.5$, is designated by τ , then from (40)

$$\tau = \frac{0.69315}{\frac{2}{3} \frac{kT}{r} \frac{r_1}{r_2} \left(1 + \frac{A\ell}{r_2} \right) n_1} \quad (41)$$

From this formula it is seen that τ is significantly smaller than T , r_1 and n_1 are larger, and that r_2 is very small, the value of τ decreasing rapidly when r_2 becomes smaller than ℓ .

As an example, formula (41) has been used to calculate the capture of atmospheric nuclei of 0.01 μ , 0.05 μ and 0.10 μ radius by cloud droplets of 10 μ radius, the concentration of the latter being 200 droplets per cm³. In setting $k = 1.3803 \times 10^{-16}$ erg °K⁻¹, $n = 1.818 \times 10^{-4}$ g cm⁻¹ sec⁻¹, $T = 293^\circ$ K, $A \approx 1$ and $\ell = 0.1\mu$ the values contained in Table 6 may be obtained.

TABLE 6

Half life of atmospheric nuclei in a cloud composed of droplets of 10 μ radius and of concentration 200 droplets per cm³

$r_2(\mu)$	0.01	0.05	0.10
$\tau(\text{hours})$	0.59	10.8	32

If all the nuclei have the same radius ($r_1 = r_2 = r$), formula (38) may be written

$$\frac{\partial}{\partial t} \left(\frac{1}{n} \right) = \frac{8}{3} \frac{kT}{n} \left(1 + A \frac{\ell}{r} \right) \quad (42)$$

Integrating this equation between the initial state (t_0, n_0) and some state (t, n), it follows that

$$\frac{1}{n} - \frac{1}{n_0} = \frac{8}{3} \frac{kT}{n} \left(1 + A \frac{\ell}{r} \right) (t - t_0) \quad (43)$$

from which it can be deduced that

$$\tau = \frac{1}{\frac{8}{3} \frac{kT}{n} \left(1 + A \frac{\ell}{r} \right) n_0} \quad (44)$$

This formula shows, for example, that it takes 312 hours for the concentration of an aerosol containing 1000 nuclei of 0.2μ radius per cm^3 to be reduced by a factor of two.

Consider now the case where formula (3) is applicable. From (20') it follows then that

$$\frac{\partial n}{\partial t} = -\frac{2}{3} \frac{kT}{n} K\ell \left(\frac{1}{r_1^2} + \frac{1}{r_2^2} \right) (r_1 + r_2) n_1 n_2 \quad (45)$$

When the aerosol is homogeneous ($r_1 = r_2 = r$), (45) may be written

$$\frac{\partial}{\partial t} \left(\frac{1}{n} \right) = \frac{8}{3} \frac{kT}{n} \frac{K\ell}{r} \quad (46)$$

from which can be successively deduced as from (42), the following formulae

$$\frac{1}{n} - \frac{1}{n_0} = \frac{8}{3} \frac{kT}{n} \frac{K\ell}{r} (t - t_0) \quad (47)$$

$$\tau = \frac{1}{\frac{8}{3} \frac{kT}{n} \frac{K\ell}{r} n_0} \quad (48)$$

Formula (48) shows, for example, that it takes 0.47 hour for the concentration of an aerosol containing 10^5 nuclei, of 0.01μ radius,

per cm^3 to be reduced by a factor of two. This result shows the importance of thermal coagulation for Aitken nuclei.

Junge (1957) used formula (38) to study the influence of thermal coagulation on the variation in the granulometry of atmospheric nuclei. The results he obtained for a continental aerosol are represented in Fig. 6.

CAPTION Figure 6. Variation with time of the average distribution of the concentration of nuclei as a function of their radius r , as a result of thermal coagulation (Junge, 1957). The curves marked 0^h , 1^h , 4^h , 24^h or 1^d , 3^d , and 7^d correspond, respectively, to the initial distribution ($t = 0$) and to the distributions after 1 hour, 4 hours, 1 day, 3 days, and 7 days.

For the small Aitken nuclei it is seen that the decrease in concentration as a function of time is rapid and that that of the large Aitken nuclei, of the large nuclei and of the giant nuclei is, on the contrary, insignificant. As a result thermal coagulation has the effect of displacing the peak of the curve of the nuclei distribution toward the large nuclei. Also as a result the life span of nuclei of radius less than $5 \times 10^{-3} \mu$ is ephemeral, because the lower limit of the radii of atmospheric nuclei is a consequence of thermal coagulation. Apparently the decrease of the total mass of Aitken nuclei is compensated by an increase in the total mass of large nuclei. As a consequence thermal coagulation has the effect of transporting Aitken nuclei onto large nuclei which explains why these nuclei are, in general, mixed nuclei.

4.3. Atmospheric scavenging

The mechanisms by which water acts as a scavenging agent in the atmosphere can be divided into two categories (see, for example, Facy, 1960 and Junge, 1963b). In the first category fall the mechanisms capable of transporting nuclei onto cloud particles and in the second the mechanisms causing the capture of nuclei situated in the path of the precipitation particles.

First we shall examine those mechanisms falling in the first category. First of all there is the thermal capture of nuclei by cloud particles which theoretically can be treated as the problem of the coagulation of aerosols, the cloud particles being considered as the nuclei. It is doubtful that the equations of Brownian motion can be applied without reservation in this case. However, it can be agreed that they are usable, without losing sight of the fact that the values obtained constitute only an order of magnitude; the example which we gave earlier (see 4.2.4.) shows this mechanism to be important for the capture of small Aitken nuclei.

Secondly, there is the capture of nuclei by cloud particles resulting from the flux of diffusing vapor which entrains nuclei toward the particles. The force exerted by the diffusing vapor transmits to them a speed proportional to the gradient of the vapor and independent of the radius of the particle. It was Facy (1955, 1958) who drew attention to this phenomenon for the case of cloud droplets. According to that author, the action of this mechanism is greater than that of Brownian motion, which is not the opinion of Deriagin and Dukhin (1957), of Severynse (1963) and of Goldsmith, Delafield and Cox (1961, 1963) who have shown theoretically and experimentally that the efficiency of this phenomenon is low within aqueous clouds. Podzimek (1965) is also of this opinion but nevertheless thinks that this phenomenon could be of importance in mixed clouds because of the increased water vapor gradient which should exist around the dendritic points in the neighborhood of the droplets.

Let us now examine the capture of atmospheric nuclei by precipitation particles, i.e., that which is usually referred to as scavenging of the atmosphere because the drops of rain and crystals of snow remove, in particular, impurities suspended in the atmosphere: industrial pollutants, radioactive debris, biological organisms, dust, etc.

Let us assume that the precipitation particles are spheres, which is, as a first approximation, acceptable in the case of drops of rain, but which is difficult to accept in the case of snow crystals. Let us also assume that each collision of a precipitation particle and a nucleus results in a capture which is certainly debatable especially if the nuclei are not wettable. The problem considered, then, is that of the aerodynamic capture of particles by spheres, a complicated problem which has been rigorously solved in only two limiting cases: the first, that of the viscous flow (Reynolds Number $Re \rightarrow 0$) and the second, that of the potential flow ($Re \rightarrow \infty$) (see, for example, Herne, 1960 and Fuchs, 1964). For a highly idealized case, Greenfield (1957) found that the scavenging of the atmosphere by rain drops is significant only for those nuclei of radius greater than 5μ . This result has been confirmed by Hess (1959) who has shown that a two-hour rain of an intensity of 1 mm per hour would scavenge 85 to 97% of the nuclei of radius about 5μ . In contrast, a rain of less than one hour of intensity less than 2 mm per hour in practice doesn't capture nuclei of radius equal to 1.5μ .

Walton and Woodcock (1963) have studied in vitro the phenomenon of scavenging by suspending drops of 0.25 mm, 0.5 mm and 1.0 mm radius in a vertical tube in which circulated, at speeds corresponding to different droplet fall speeds, a homogeneous aerosol of particles of methylene blue of radius equal to 2.5μ or to 1.25μ . These authors compared their results with the values calculated by Fonda and Herne (Herne, 1960) assuming the flow at potential speed around a sphere. It was thus established that,

for a drop of given radius, the capture coefficient* E decreases rapidly when the particle radius decreases. The variation is practically the same whatever the radius of the droplet: E has a value of about 0.7 for $r = 5\mu$, 0.3 for $r = 2.5\mu$ and 0.01 for $r = 1\mu$.

* The capture coefficient is the ratio between the number of particles captured by the sphere and the number of particles contained in the volume swept out by the sphere. This coefficient can be greater than one, because particles not in this volume can be aspirated in the trail and collide with the sphere.

Starr and Mason (1966) have studied in vitro the capture, by water drops of radius varying between 0.1 mm and 1.0 mm, of lycoperdon spores, of spores of a blight (*ustilago nuda*) and of grains of a pollen (paper mulberry) of which the mean radii \bar{r} were respectively 2.25μ , 2.6μ and 6.4μ . For the two kinds of spores, the capture coefficient E increased rapidly as the radius of the drops increased, passed through a maximum when the radius was about 0.4 mm, then decreased more slowly. For the lycoperdon spores the value of E is about 0.05 when the radius of the drops is 0.1 mm and 1.0 mm and in the vicinity of 0.15 when the radius is 0.4 mm. For the *ustilago nuda* spores, these values are, respectively, near 0.10 and 0.20. These results correspond relatively well with the values calculated under the assumption of potential flow for the descending part of the flow where this assumption is valid. For the paper mulberry grains, the capture coefficient is about 0.50 when the radius of the drops is 0.1 mm; it then increases slowly passing through a poorly defined maximum at about 0.70 when that radius is close to 0.4 mm, then decreases very slowly and is still 0.60 when the radius is 1.0 mm. These results correspond well with the calculations of Langmuir derived from an interpolation between viscous flow and potential flow (see, for example, Herne, 1960).

Engelmann (1965) studied in vivo the phenomenon of scavenging of the atmosphere by causing an artificial rain, of drop radius between 0.2 mm and 0.8 mm, to fall in a horizontal plane of particles of zinc sulfide of radius varying between 1.8μ and 7.0μ . In this manner it was found: that the capture coefficient E is greater than one when the particle radius exceeds 6.5μ , whatever the radius of the drops; that this coefficient is a maximum when the radius of the drops is near 0.2 mm and that it passes through an ill defined minimum when this radius is close to 0.4 mm; that this coefficient increases for a given radius of the drops when the dimensions of the particles increase, the increase becoming rapid when the radius of the particles exceeds 5μ .

The results of these studies, very different from certain points of view, nevertheless agree on one point - knowledge that the scavenging of the atmosphere by rain drops becomes significant only for nuclei of radius greater than several microns. This phenomenon thus has little effect on the major portion of the atmospheric aerosol and affects only the giant nuclei.

Little is known about the scavenging of the atmosphere by solid precipitation. Starr and Mason (1966) studied the capture of lycoperdon spores ($\bar{r} = 2.25\mu$) and pollen grains of paper mulberry ($\bar{r} = 6.4\mu$) through pieces of tissue paper cut in the form of a circle, of a hexagon, or of a star in order to simulate snow crystals. The capture coefficients deduced from the number of particles captured by the lower face of these simulated crystals are very low and less than 0.05. Nevertheless it should be noted that the upper face of these pieces of paper captured a number of particles essentially equal to that which was observed on the lower face which seems to indicate that the effect of aspiration is important in the case of snow crystals.

Further observations are necessary to clear up the problem of the scavenging of the atmosphere, a problem of importance not only in meteorology but also in industry, for example in the suppression of dust by aspiration.

4.4. Remarks

According to Junge (1961), the vertical variation of the concentration n of the nuclei of a homogeneous aerosol in the vertical plane can be calculated, in a permanent regime ($\frac{\partial n}{\partial t} = 0$), by neglecting the variation of D and of v_1 with z by means of the differential equation

$$\frac{\partial n}{\partial t} = D \frac{d^2 n}{dz^2} - v_1 \frac{dn}{dz} - bn^2 - an = 0 \quad (49)$$

where D is the coefficient of turbulent diffusion of the nuclei, z the altitude, v_1 the terminal speed of the nuclei, b the coefficient of thermal coagulation and a the coefficient of scavenging.

The different terms of equation (49) represent: the first, the effect of turbulent diffusion (see Eq. 13); the second, the effect of sedimentation (see Eq. 13); the third, the effect of thermal coagulation (see Eq. 36) and the fourth, the effect of scavenging. This equation does not take into account that which could be termed the effect of condensation resulting from the formation of atmospheric droplets around condensation nuclei, a significant factor for large nuclei.

For Aitken nuclei: the effect of turbulent diffusion is important; the effect of coagulation equally so, especially for the small nuclei; the effect of scavenging is weak and the effect of sedimentation is negligible. Equation (49) then takes the form

$$D \frac{d^2 n}{dz^2} - bn^2 \approx 0 \quad (50)$$

an equation utilized by Selezneva (1966) to study the vertical distribution of Aitken nuclei.

For large nuclei: the effect of turbulent diffusion is important; the effect of scavenging begins to become significant; the effect of coagulation practically disappears and the effect of sedimentation remains negligible. Equation (49) then takes the form

$$D \frac{d^2 n}{dz^2} - an \approx 0 \quad (51)$$

an equation used by Junge (1961) to study the vertical distribution of Aitken nuclei above 5 km, in making the debatable assumption that the effect of scavenging is more important than the effect of coagulation in this part of the atmosphere.

For giant nuclei: the effect of turbulent diffusion is important for the small nuclei within this category but decreases rapidly as the size increases; the effect of sedimentation undergoes an inverse variation; the effect of scavenging increases rapidly with the size of the nuclei and becomes important when the radius exceeds about 5 μ ; the effect of coagulation is negligible. Equation (49) then takes the form

$$D \frac{d^2 n}{dz^2} - v_1 \frac{dn}{dz} - an \approx 0 \quad (52)$$

If the effect of scavenging is disregarded, this equation may be written

$$D \frac{d^2 n}{dz^2} - v_1 \frac{dn}{dz} \approx 0 \quad (53)$$

an equation used by Junge (1957) to study the effect of turbulence on the sedimentation of nuclei (see 4.1.3.).

We shall no longer stress these equations which not only strongly idealize the phenomena but which are also difficult to utilize, lacking accurate data on the value of the coefficients which they contain, especially in regard to that concerning the effect of scavenging.

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